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Fingering Flow Patterns of Thermosolutal Convection in Rectangular Enclosures

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FINGERING FLOW PATTERNS OF THERMOSOLUTAL CONVECTION IN RECTANGULAR ENCLOSURES

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SUMMARY

Convection in rectangular enclosures with combined horizontal temperature and concentration gradients is studied. An electrochemical system is employed to impose the concentration gradients. Due to a large difference between the thermal and solutal diffusion rates the flow possesses double-diffusive characteristics. Very complex fingering flow patterns are observed around the two-phase interfaces. The main objective of the present work is to obtain more information on the flows near the two vertical phase interfaces under various parametric conditions. The fingering flows near the crystal could be one of the most important factors to cause the crystal imperfections.

The ranges of the parameters studied are Sc (Schmidt number) = 2200 to 2400, Pr (Prandtl number) = 4.0 to 7.0, Gr_T (Grashof number) = 2.50×10^6 to 5.01×10^7, Ar (aspect ratio) = 0.61 to 3.0, and N (buoyancy ratio) = 0.05 to 54.8.

E-4057-1

INTRODUCTION

The growth of crystal is a coupled process of heat and mass transfer, fluid flow, phase transformations, and chemical reactions. Although the technology of crystal growth is well grounded in physical chemistry, the treatment of the transport phenomena has been relatively rudimentary and empirical. Recently, it was recognized that convection, as a result of temperature and concentration gradients, occurs in many aspects of materials processing including solidification, oxidation of surface material, and crystal growth.

Thermosolutal convection is very important in crystal growth processes (ref. 1). In many crystal growth techniques (e.g., horizontal Bridgman) the fluid phase is subjected to horizontal temperature and concentration gradients. Although several investigators in the past studied thermosolutal bulk convection in enclosures with combined horizontal temperature and concentration gradients (refs. 2 to 4), extensive and systematic work is still needed to

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understand convection near the two vertical metal plates in enclosures with combined driving forces.

In almost all the commonly used crystal growth techniques the parent phase is a fluid. As in most nonequilibrium processes involving a fluid, convection plays a dominant role in that it affects the fluid-phase composition and temperature at the phase interface. The fluid motion in many cases is determined by the locally generated motion. The flows are due either to the effects of temperature and/or concentration gradients in a body-force field. In some crystal-growth techniques, the completely confined fluid phase is subject to horizontal temperature and concentration gradients. To gain insights on such flows, an experimental program was initiated to study flows near the phase interfaces in rectangular enclosures with horizontal temperature and concentration gradients.

Despite the relevance of double-diffusive convection near the fluid-solid interface, little work has been done in this area. The objective of this study is to investigate double-diffusive convection induced by horizontal temperature and concentration gradients in enclosures. Attention is focused on the flow structure of convecting regions near the metal plates.

EXPERIMENTAL DESIGN

Dimensionless Parameters

Based on the basic differential equations for natural convection in an enclosure with thermal and solutal buoyancy forces (ref. 1), it can be shown that the following dimensionless parameters are important in the present work:

$$\text{thermal Grashof number, } Gr_T = \frac{g\beta\ \Delta T H^3}{\nu^2}$$

$$\text{Prandtl number, } Pr = \frac{\nu}{\alpha}$$

$$\text{Schmidt number, } Sc = \frac{\nu}{D}$$

$$\text{buoyancy ratio, } N = \frac{\bar{\beta}\ \Delta C}{\beta\ \Delta T}$$

$$\text{aspect ratio, } Ar = \frac{H}{L}$$

where g is the gravitational acceleration, ν is the fluid kinematic viscosity, α the thermal diffusivity, and D the diffusion coefficient. The height and width of the enclosure are H and L, respectively. ΔT and ΔC are the imposed temperature and concentration differences, respectively, between the two vertical walls separated by L. Density variation due to temperature is represented by the volumetric thermal expansion coefficient β, and that due to concentration by the volumetric solutal expansion coefficient $\bar{\beta}$. Instead of

the (Gr_T, N) combination the (Gr_T,Gr_S) combination is sometimes used, where

Gr_S is defined as $Gr_S = \frac{g\bar{\beta}\,\Delta CH^3}{\nu^2} = N\,Gr_T$, solutal Grashof number.

With the present system the ranges of the dimensionless parameters covered for thermal and solutal cases are Pr = 4.0 to 7.0, $Gr_T = 2.50 \times 10^6$ to 5.01×10^7, Ar = 0.61 to 3.0, N = 0.05 to 54.8, and Sc = 2200 to 2400. All fluid properties are evaluated at the average temperature of the hot and cold walls and at the average concentration of the bulk solution.

Test Apparatus

A sketch of the experimental system is presented in figure 1. The test cell is a rectangular enclosure formed with four insulating plexiglas plates and two copper plates (both thermally and electrically conductive). The width, L, is 7.7 cm and the height is variable so that a range of aspect ratios can be covered. The depth of the enclosure is relatively large (23.9 cm). The two copper vertical end walls that are 0.7 cm thick.

An electrical heating mat is bonded on the back of one of the copper plates and a circulating water system cools the other copper plate, so that uniform temperatures can be imposed on the copper surfaces. The plexiglas surfaces are wrapped with 5 cm thick fiberglass insulation to ensure an adiabatic boundary condition. There are three thermocouples embedded in each copper plate to measure the uniformity of the temperature over the entire surface. A thermocouple probe with a two-dimensional traversing mechanism is inserted into the test section to measure the temperature distribution inside the enclosure. A Leeds and Northrup millivolt potentiometer is used to measure the thermoelectric emf from the copper-constantan thermocouples.

The test fluid is a copper-sulphate-acid solution ($CuSO_4 + H_2SO_4 + H_2O$). When a voltage is applied to the electrodes, copper dissolves into the solution at the anode and is deposited at the cathode. As a result, the density of the fluid near the cathode (anode) becomes lower (higher) than that of the bulk of the solution. The migration of the cupric ions in the electrical field is eliminated by adding sulphuric acid to the solution, which acts as a supporting electrolyte, and thus the transport of the cupric ions in the cell is controlled only by diffusion and convection. In the experiment the concentration of $CuSO_4$ varies from 0.05 to 0.10 M. The acidity of the solution is kept constant at 1.5 M H_2SO_4. The physical properties of the solution are taken from Wilke et al (ref. 5).

The auxiliary system consists of a dc power supply, instruments to measure the total current and potential in the cell, and a variable resistance to control the current.

Test Procedure

Although the temperatures of the copper walls can be easily measured by thermocouples, the concentration levels at the walls cannot be so easily determined. One relatively simple way to specify the concentration level at the

cathode wall in the present system is to adjust the cell potential in such a way that the limiting current is obtained. Under the limiting-current condition the ion concentration at the cathode surface is zero, in other words the change in concentration across the solutal boundary layer along the cathode is Cb, where Cb is the concentration level in the bulk fluid. As convection develops in the cell, the concentration level becomes nonuniform in the bulk region, and only on the average the bulk concentration level remains constant. Two types of tests have been conducted. In the first type of tests, the electrochemical system is started after the thermal convection becomes steady. Due to electrolysis the copper wall surfaces become rough slowly with time, the duration of each run is limited to a maximum 5 hr after the electrochemical system is started. However, the time required to attain steady solutal convection is far longer than 5 hr. In the second type of tests, the system is started in a different way, the electrochemical system is started first for 4.5 hr and then a temperature gradient is imposed. Since the limiting current has been found to change gradually with time as thermal convection develops in the cell, the concentration boundary conditions become less well defined. To visualize local flow structures, laser light and dye are employed. The optical technique is shadowgraphs.

RESULTS AND DISCUSSION

The First Type of Tests

It could be anticipated that the flow structures for combined driving force cases are more complex than those in purely thermal or solutal cases (ref. 2). The sketch of typical layered flow pattern observed with dye tracers for a case is given in figure 2. Using optical visualization techniques, we have found that secondary motions appear near each vertical wall. Further, in cells near the hot anode the flows are counterclockwise.

Because the thermal convection dominates in the bulk flow in the present system, near the hot anode the fluid moves upward along the wall to the interface where some of the heavier fluid entrained from the solutal layer turns downward. The remainder of the fluid proceeds toward the other end along the interface. Similar behavior occurs near the cold cathode. The result is layered patterns with secondary cells near the electrodes. The secondary cells are a result of the double diffusion because as the flow turns near the corner of the interface and the copper plate the response to the temperature change is rapid but the concentration does not change appreciably. Thus the entrained fluid that leaves the thermal boundary layer has a buoyancy force opposite to the main flow and it moves in a counter direction to form the chaotic turbulent fingering cell.

Shown in figures 3 and 4 are some salt fingers near the copper plates. The salt fingers for the present cases are due to the fact that fluid parcels heated at the anode rise in a decreasing ambient density environment (induced by the horizontal concentration gradient) which limits their ascent, the parcel then turns around and becomes salt finger. What is called "salt fingering" occurs as a result of the interaction between a stable temperature gradient (ref. 6) and an unstable concentration gradient. The appearance of salt fingers can be modified by geometrical constraints as a comparison of figures 3(a) to (c) indicates. For a fixed solutal Grashof number and aspect ratio when the

buoyancy ratio, N is reduced (thermal Grashof number increase), the salt fingers occur at higher positions along the anode for the opposing cases.

The Second Type of Tests

The electrochemical system is started first for 4.5 hr and then a temperature gradient is imposed. When fluid with a vertical concentration gradient induced by a horizontal concentration gradient in an enclosure is heated and cooled from the sides, the resultant motion consists of a series of layers which propagate away from the heated and cooled walls (fig. 5). The motion is driven by the rise (drop) of the warmed (cooled) fluid parcels near the wall. Because the diffusion coefficient of $CuSO_4$ is much less than that of heat, the parcels retain almost all their concentration as they rise (drop) in a continually decreasing ambient density field. Their positive (negative) buoyancy force also continually decreases and a level is reached beyond which no further vertical motion is possible; the parcel then turns horizontally into the interior. Many horizontally oriented layers are thereby set up.

Similar phenomena can be achieved by the other way which is a smooth vertical concentration gradient heating or cooling from a side wall directly (ref. 7). A hallmark of this double-diffusive convection is the transformation of a smooth vertical concentration gradient into a series of layers separated by very thin interfaces. Relatively large variations in concentrations exist across these interfaces. Double-diffusive intrusions form within a matter of minutes after a temperature gradient is imposed. Figure 5 shows shadowgraphs of an experiment which has been heated and cooled from the sides for a few minutes. The convection cells or rolls (look like thick fingers, see fig. 5(d)) spring up along the heated and cooled wall. This may indicate a stability phenomenon (ref. 7). The cells are approximately equal size. They grow in the lateral direction.

CONCLUSION

The results of the present experiments indicate that for the range of aspect ratios, buoyancy ratios, and Grashof and Prandtl numbers covered:

1. In the first type of tests, it is found that the interaction between the temperature and concentration field may cause secondary fingering cells near two end walls in the layered flow structure.

2. In the first type of tests, if fingers appear along the copper plate, their positions are dependent on N for a fixed Ar and Grs.

3. In the second type of tests, a series of layers separated by very thin interfaces appeared in the bulk flow. The layer thickness was much thinner than those in the first type of tests. The flow becomes unstable and cellular convection results, which is clearly visible in shadowgraphs.

The layered structures, salt fingers, secondary cells, and the sideways diffusive instability all may affect the crystallization. Thus, there is a need for considerable more research on problems of this type.

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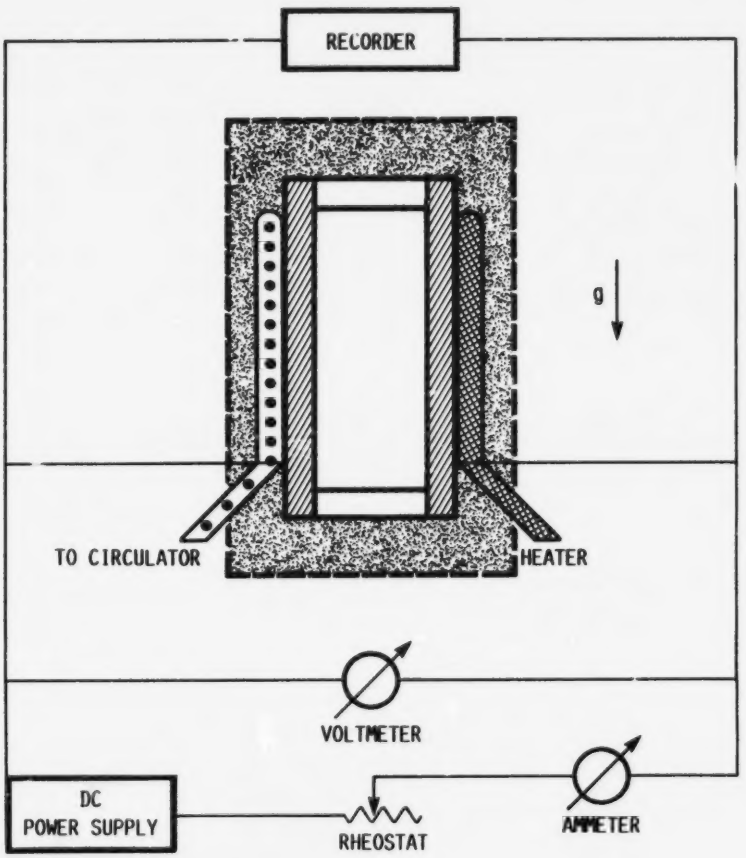
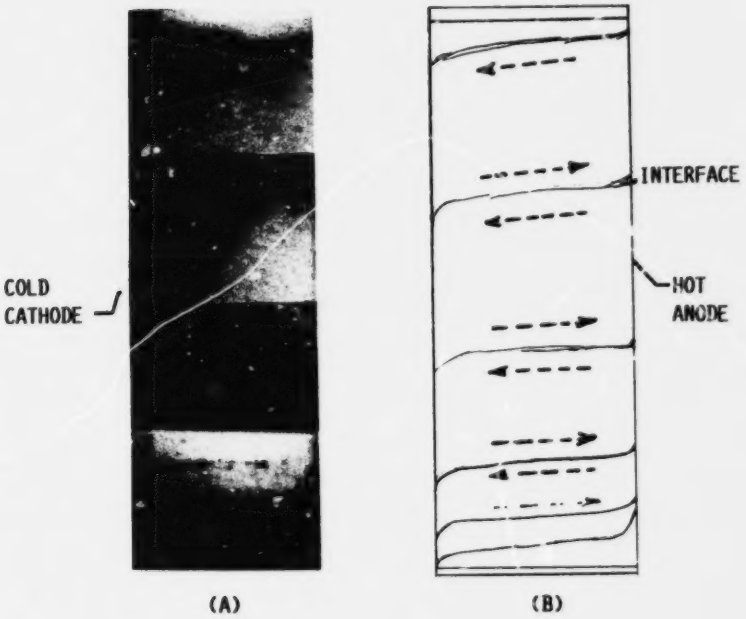
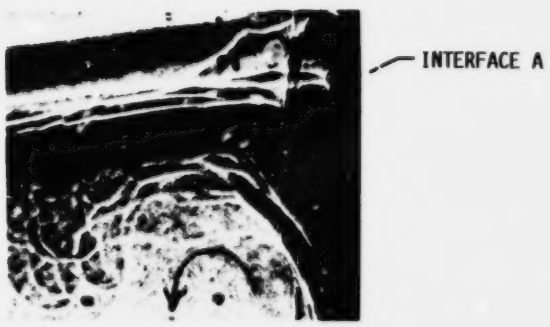


FIGURE 1. - ILLUSTRATION OF TEST CELL AND ELECTRIC CIRCUIT.





(A) BULK FLOW PATTERNS.
(B) BULK FLOW PATTERNS.
(C) SECONDARY FINGERING CELL NEAR ANODE IN THE MIDDLE LAYER.

FIGURE 2. - FLOW PATTERNS ($Ar = 3$, $Gr_T = 5.01 \times 10^7$, $N = 0.30$ FOR AN OPPOSING CASE).

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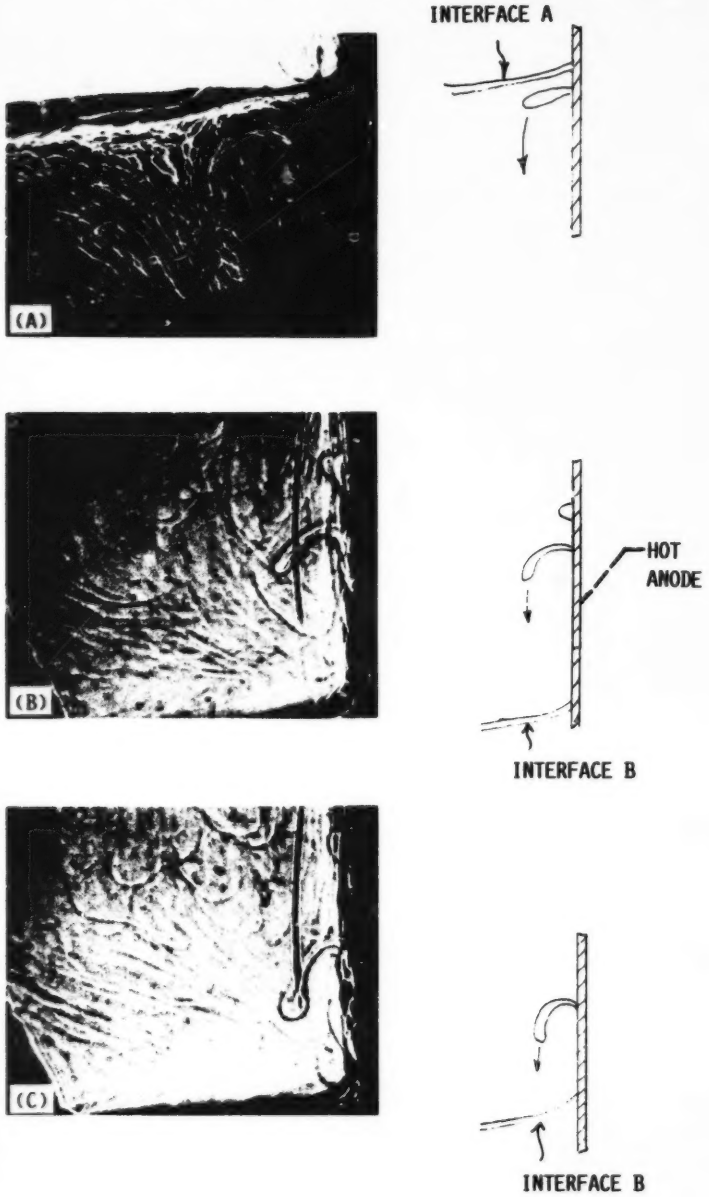
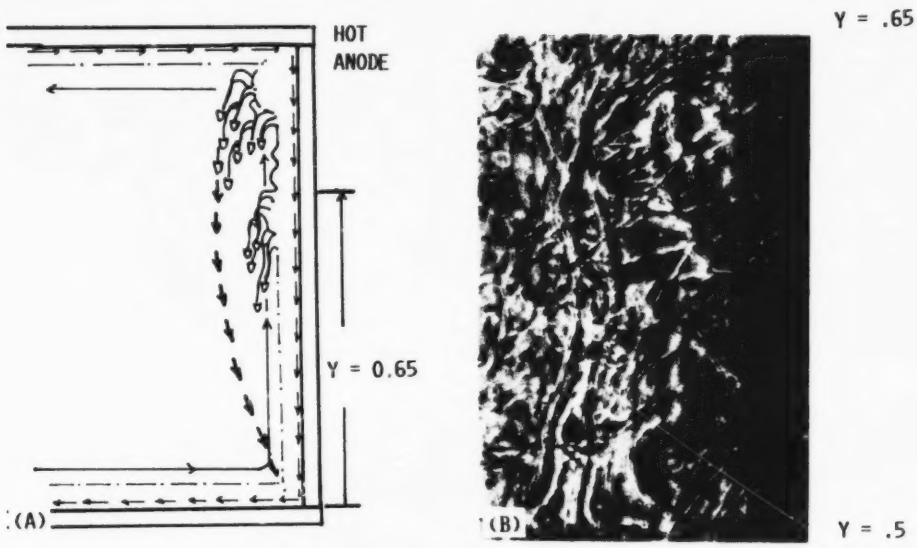


(A) N = 0.05.
(B) N = 0.15.
(C) N = 0.30.

FIGURE 3. - FINGERING FLOW PATTERNS NEAR THE HOT ANODE (Ar = 3, $Gr_S = 1.52 \times 10^7$). POSITIONS OF INTERFACE A IS HIGHER THAN THE POSITION OF INTERFACE.



(A) SKETCH OF FLOW PATTERNS NEAR THE HOT ANODE.
(B) FINGERING FLOW PATTERNS NEAR THE ANODE AT Y = 0.5 TO 0.65.

FIGURE 4. - FLOW PATTERNS (Ar = 0.61, N = 6.25, $Gr_S = 4.2 \times 10^7$).

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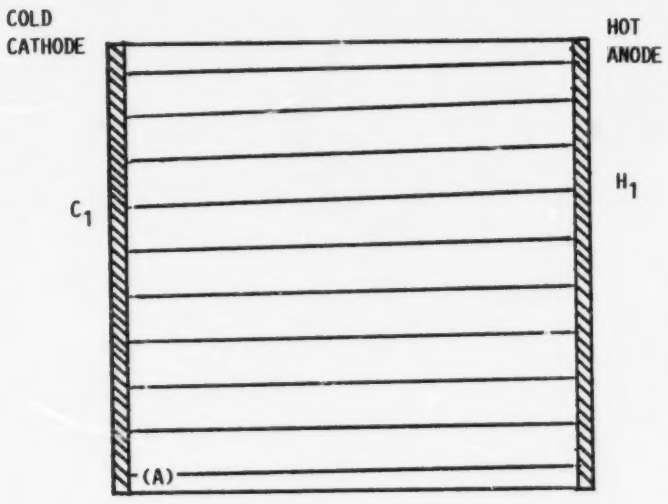
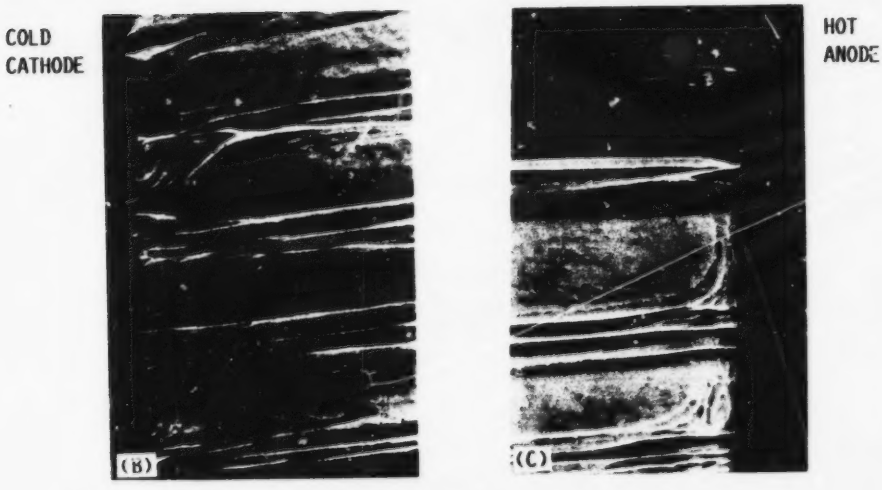
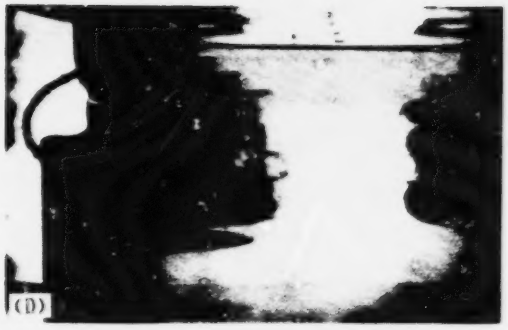




(A) A SERIES OF LAYERS APPEARS IN THE FLOW.
(B) LEFT SIDE OF THE TEST CELL.
(C) RIGHT SIDE OF THE TEST CELL.

FIGURE 5. - FLOW PATTERNS (Ar = 0.96, N = 54.8, $Gr_T = 2.5 \times 10^6$).



(D) AFTER THE THERMAL GRADIENT IS IMPOSED.
(E) UPPER SIDE OF THE TEST CELL.
(F) LOWER SIDE OF THE TEST CELL.

FIGURE 5. - CONCLUDED.

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16. Abstract

Convection in rectangular enclosures with combined horizontal temperature and concentration gradients is studied. An electrochemical system is employed to impose the concentration gradients. Due to a large difference between the thermal and solutal diffusion rates the flow possesses double-diffusive characteristics. Very complex fingering flow patterns are observed around the two-phase interfaces. The main objective of the present work is to obtain more information on the flows near the two vertical phase interfaces under various parametric conditions. The fingering flows near the crystal could be one of the most important factors to cause the crystal imperfections. The ranges of the parameters studied are Sc (Schmidt number) = 2200 to 2400, Pr (Prandtl number) = 4.0 to 7.0, Gr_T (Grashof number) = 2.50×10^6 to 5.01×10^7, Ar (aspect ratio) = 0.61 to 3.0, and N (buoyancy ratio) = 0.05 to 54.8.

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